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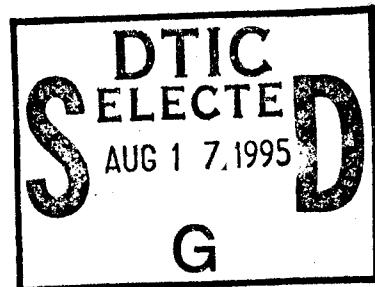
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UNITED STATES ATOMIC ENERGY COMMISSION

A PROBE-TYPE ALPHA DETECTOR FOR THE  
MEASUREMENT OF POLONIUM SOLUTIONS

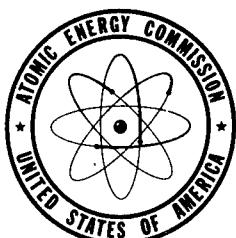
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LOS ALAMOS SCIENTIFIC LABORATORY  
of the  
UNIVERSITY OF CALIFORNIA

Report written:  
March, 1953

LA-1523

THE PREPARATION AND REDUCTION OF  
POLONIUM DIOXIDE  
(Final Report on Project Authorization CMR-3-8)

Work done by:

George Moulton  
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Report written by:

George Moulton  
John Farr  
Dwayne Vier

## ABSTRACT

The preparation and reduction of polonium dioxide,  $\text{PoO}_2$ , is described. The light yellow-orange colored dioxide is formed by the reaction of polonium metal and dry oxygen at a temperature of  $250^\circ\text{C}$  and an oxygen pressure of 40 mm of Hg. Hydrogen at a pressure of 40 mm of Hg quantitatively reduces the oxide to the metal at a temperature of  $200^\circ\text{C}$ .

Work performed under Contract No. W-7405-Eng-36

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### 1. Introduction

The usual analysis of a solution for polonium content is effected by evaporating a known aliquot of the solution onto a metal foil and counting the foil, for alpha emission, in a methane proportional counter. Although this method gives great accuracy, it is time-consuming and frequently, e.g., in the series of polonium recovery processes,<sup>1</sup> the results of the analysis must be awaited before the next step of the operation in progress can be performed.

The probe-type alpha detector described in this paper was designed and constructed to fulfill the need for an instrument to measure the polonium content of solutions rapidly, conveniently, and continuously.

### 2. Design and Construction

An ionization-chamber mode of operation, rather than the Geiger-Mueller principle, was chosen for the probe detector for the following reasons: 1. Relatively low voltages are required ( $\sim 150$  volts as compared to 800-1000 volts necessary for a Geiger tube). 2. The ion-chamber response is quite insensitive to changes in applied voltage over a wide range. 3. The ion-chamber current may be amplified readily, by use of an electrometer circuit, to give a direct reading on a meter. 4. The ion chamber responds almost exclusively to alphas derived from the solution in the immediate neighborhood of the probe window and is relatively insensitive to gammas from polonium precipitates or from electroplated deposits of polonium which might be present in the vessel containing the polonium solution. Since the measurement desired is a determination of the polonium in solution, the detector chosen should be relatively insensitive to gamma radiation from polonium in the vicinity of the detector which is not in solution.

#### 2.1 Probe Design and Construction

There are several general design considerations for the probe-type alpha detector.

1. A high resistance for the ionization chamber under non-ionizing conditions.
2. Sufficient ion-chamber volume (or length of the ion chamber) so that the entering alpha particles are largely absorbed, with consequent ionization, in the ion-chamber gas.
3. A very thin, but relatively rugged, window to separate the ion chamber from the solution being measured.

The actual dimensions of the probe are quite arbitrary and may be varied to meet particular requirements.

2.1.1 Construction Precautions. Certain general precautions should be observed in the assembly of the probe.

- a. Because of the high input resistances of the amplifier, it is extremely

important that the resistance of the probe itself, under non-ionizing conditions, be greater than 50,000 megohms. This means that all soldered joints, as well as all component parts, must be washed free from rosin, oil, or grease films and then thoroughly dried before assembly. It is a wise precaution to measure the probe resistance before the probe window is finally attached in order to determine whether additional cleaning is necessary.

b. The material comprising the outside of the probe must be chemically inert so that measurements may be made in acidic or basic solutions.

c. The probe window must be thin enough to be relatively transparent to polonium alphas, but still possess sufficient mechanical strength to withstand normal usage without rupturing.

2.1.2 The Fabrication of a Probe. Mica was the material finally chosen for the probe window. Several preliminary probe models were constructed with very thin windows of glass sealed to pieces of glass tubing. However, the glass windows proved to be too fragile. In addition, it was difficult to decontaminate the glass completely after immersion in polonium solutions.

The final probe design is illustrated by the cross-sectional drawing of Fig. 1. Figure 2 is a photograph of the completed probe, and of a partially completed probe assembly. The probe was fabricated in the following manner.

a. A 4" length of 0.10" brass rod was soldered to the center lug of an amphenol connector (No. 83-798).

b. The connector was then soldered to one end of the brass tube.

c. The lucite plug was next inserted in the chamber to center and support the brass rod.

d. The outside of the brass tube (and the end of the tube) was coated with Apiezon W cement. A small flame was used to soften the cement.

e. A small sheet of mica ( $1.59 \text{ mg/cm}^2$ ) was cemented to the bottom of the brass tube and then trimmed with a sharp razor blade. Additional cement was then applied so as to overflow and cover the edges of the mica (as shown in Figs. 1 and 2). The additional cement prevents possible splitting of the mica which could result in solution being absorbed between the mica planes.

In operation, the brass tube serves as the anode and the center brass rod serves as the cathode of the ionization chamber. The probe is filled with air, at atmospheric pressure, to serve as the ionizing gas.

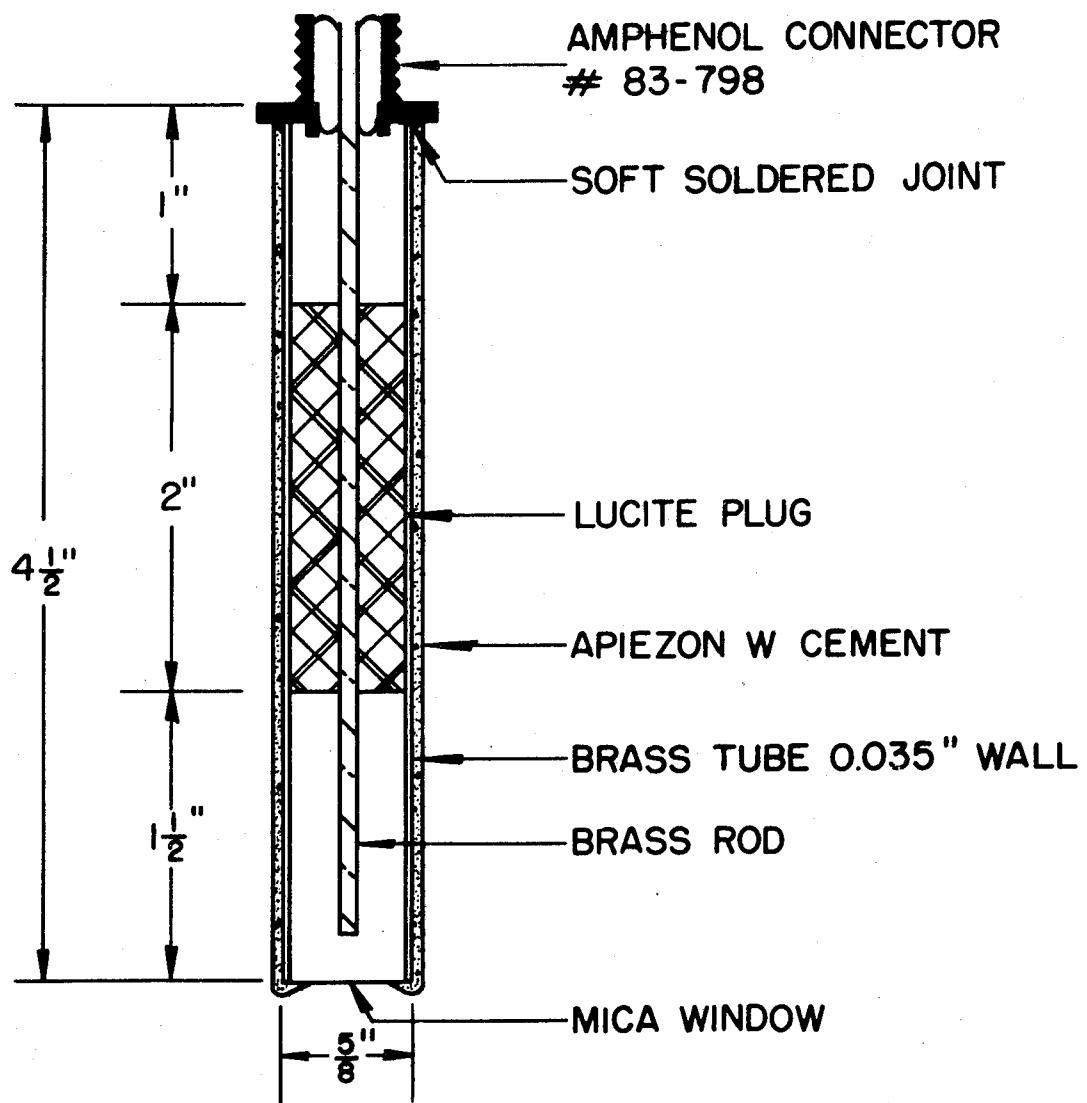


Fig. 1. Drawing of probe.

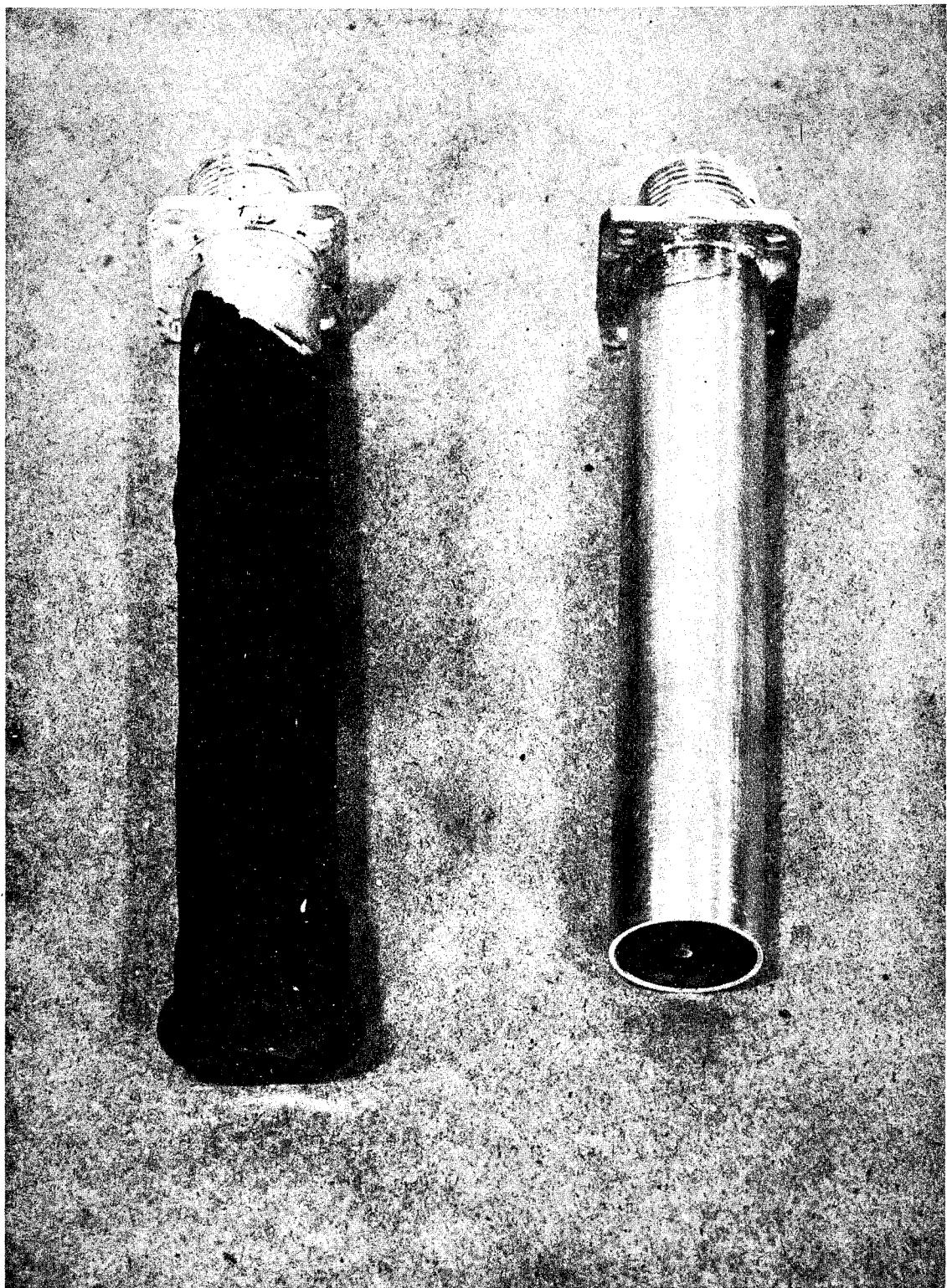


Fig. 2. Photograph of the completed probe and of a partially completed probe.

## 2.2 The Electrometer Amplifier

The electrometer amplifier is a modified version of the low-drift electrometer circuit developed by F. M. Glass.<sup>2</sup> This circuit, as modified, has proved very satisfactory. It is completely a-c powered and supplies the 150 volts d-c required for the ionization chamber, as well as the necessary amplifier voltages. The electrometer is insensitive to line-voltage fluctuations as great as 20 per cent, and has a zero drift of only 1/2 per cent per hour. Full scale deflection of the meter is obtained with 0.3 volt input so that full scale deflection, on the most sensitive range, is obtained with a current of  $6 \times 10^{-12}$  amperes.

The modified electrometer circuit is shown in Fig. 3. Noteworthy features of the circuit are described below.

a. The ganged switch ( $S_3$  of Fig. 3) was included to enhance the versatility of the instrument. This switch permits the use of an additional 5803 tube directly over the ionization probe so that the probe may be operated at distances up to 100 feet from the electrometer without increasing the response time of the instrument.

b. Since the probe operates at 150 volts above ground potential the grounded shield of the coaxial cable, which was used to connect the amplifier input and the probe, was not connected at the probe end of the cable. Instead, a separate wire connection was used to connect the amplifier chassis and the probe.

c. The top of the probe was covered with plastic tape in order to protect personnel from the 150 volt probe potential.

A photograph showing the electrometer and probe in operation is included with this paper (Fig. 4).

## 3. Calibration

The response of the electrometer is linear and varies directly with the voltage developed across the range resistors. This voltage is directly proportional to the current flowing in the range resistor circuit and this current is directly proportional to the alpha activity of the measured solution. In principle, therefore, a knowledge of the true values of the range resistances plus one calibration point for one concentration of activity is sufficient for full calibration of the instrument and probe. Unfortunately, resistors of the order of 100 megohms or more are manufactured only to tolerances of  $\pm 15\%$ . In consequence, the actual instrument calibration required either an accurate measurement of each range resistor, plus one measurement of a known polonium solution, or the calibration of each range setting against known polonium solutions. Since accurate measurements of such high resistances are quite difficult, the second method was used.

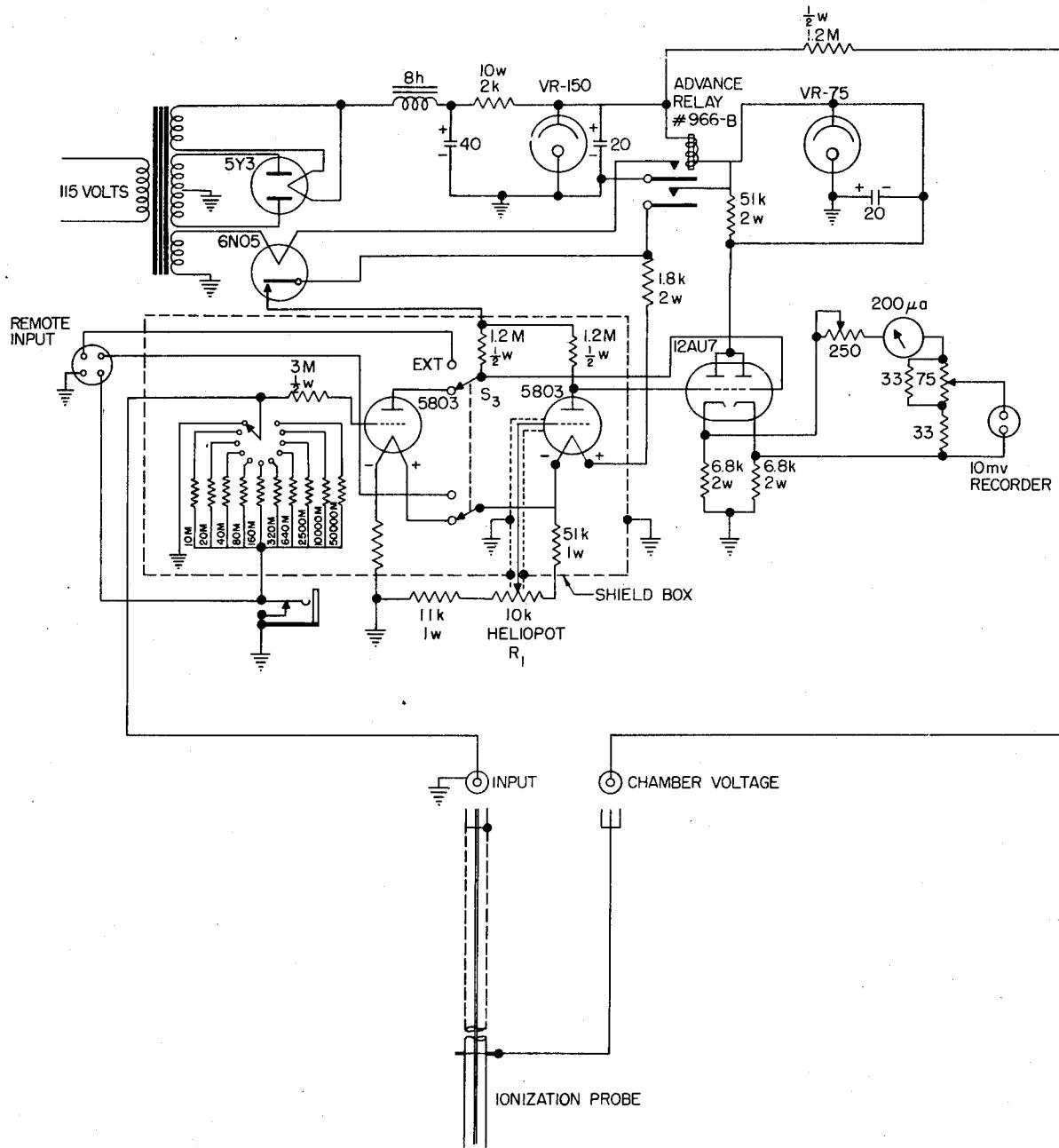


Fig. 3. The electrometer circuit.

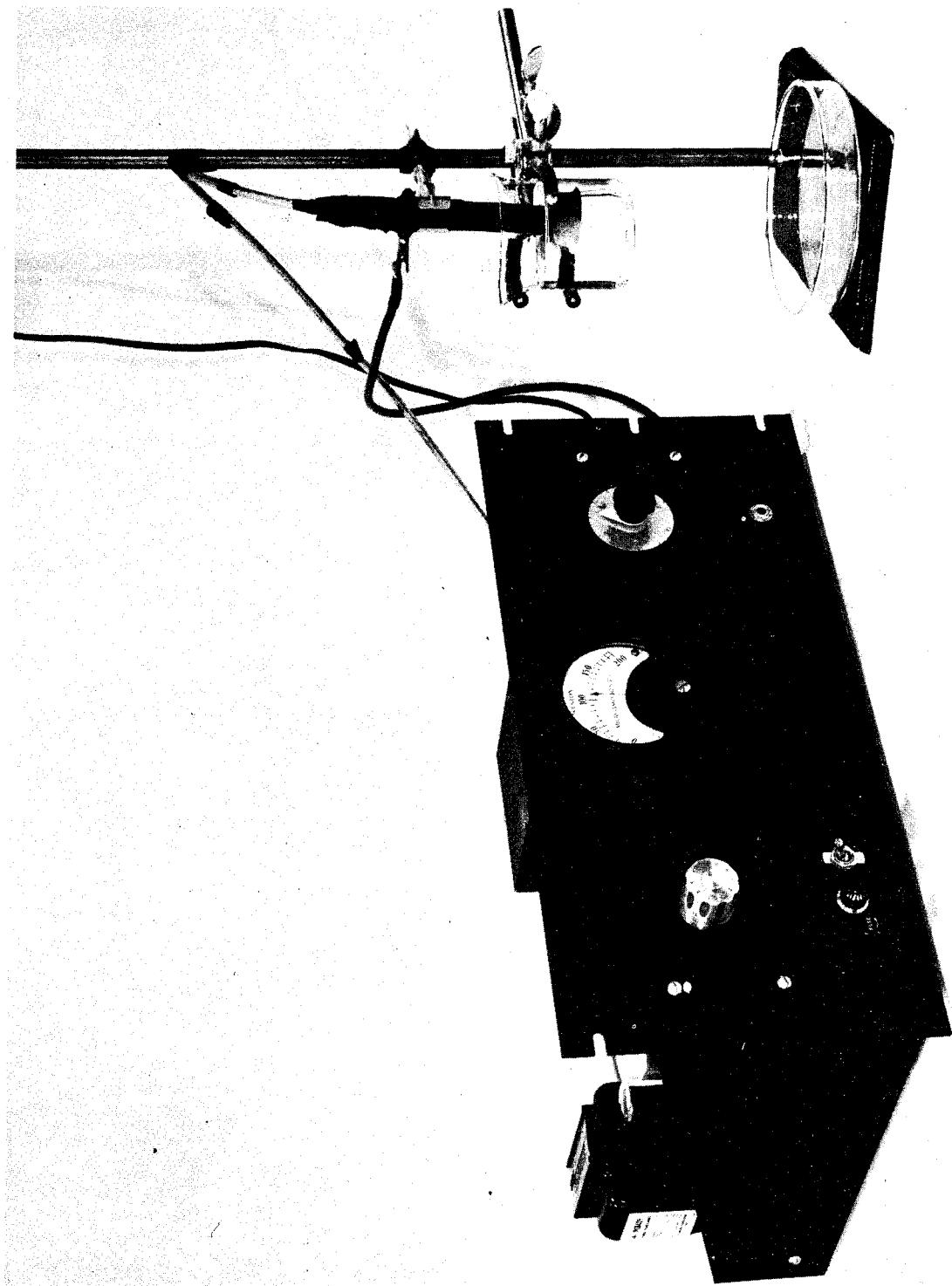


Fig. 4. Photograph of the electrometer and probe.

Ten different 1.5  $\text{N}$   $\text{HNO}_3$  solutions, containing known concentrations of polonium, were prepared. The various ranges of the amplifier were then calibrated by direct measurement of these solutions. Because of the overlapping of ranges, it was possible to obtain measurements for at least three range settings with each of the polonium solutions. Data typical of the calibration measurements are presented in Fig. 5. From these measurements, a meter-factor (millicuries of polonium per milliliter of solution per unit meter reading) was computed for each range setting. Table 1 is a tabulation of each range ratio and the corresponding meter factor computed from the solution measurements.

TABLE 1  
RATIOS AND FACTORS FOR VARIOUS RANGES

<u>Ranges</u>	<u>Ratio</u>	<u>Range</u>	<u>Meter Factor</u>	<u>Ranges</u>	<u>Ratio</u>	<u>Range</u>	<u>Meter Factor</u>
		10	0.00017	6:5	2.00	5	0.0535
10:9	4.58	9	0.00076	5:4	2.05	4	0.110
9:8	4.48	8	0.00341	4:3	2.00	3	0.219
8:7	3.52	7	0.0120	3:2	2.00	2	0.439
7:6	2.23	6	0.0268	2:1	2.00	1	0.878

It should be noted that extensive overlapping of ranges was deliberately selected so that readings near midscale are possible for any expected polonium concentration. This arrangement permits greater accuracy in the meter reading and allows a check on the instrument. With the amplifier and probe as designed, polonium concentrations from 175 mc/ml down to less than 0.01 mc/ml can be determined. The range can be readily extended if desired. All that is required is a change in the range resistance values. For example, the use of a 1 megohm range resistance will permit determination of polonium concentrations as high as 1.75 curies/ml.

#### 4. Procedure for Operation

The voltage plateau of the ionization chamber is illustrated in Fig. 6. As indicated, a value of 150 volts was selected for the operating voltage.

The electrometer amplifier should be allowed a minimum warm-up time of 30 min. The meter is then zeroed by first setting the range switch on zero position and adjusting resistance  $R_1$  (Fig. 3). The end of the probe is submerged about 1/2" below the surface of the solution, and the range switch is then moved progressively up from position 1 to 2, 3, 4 . . . etc., until a midscale reading is obtained. Because of the limited range of alpha

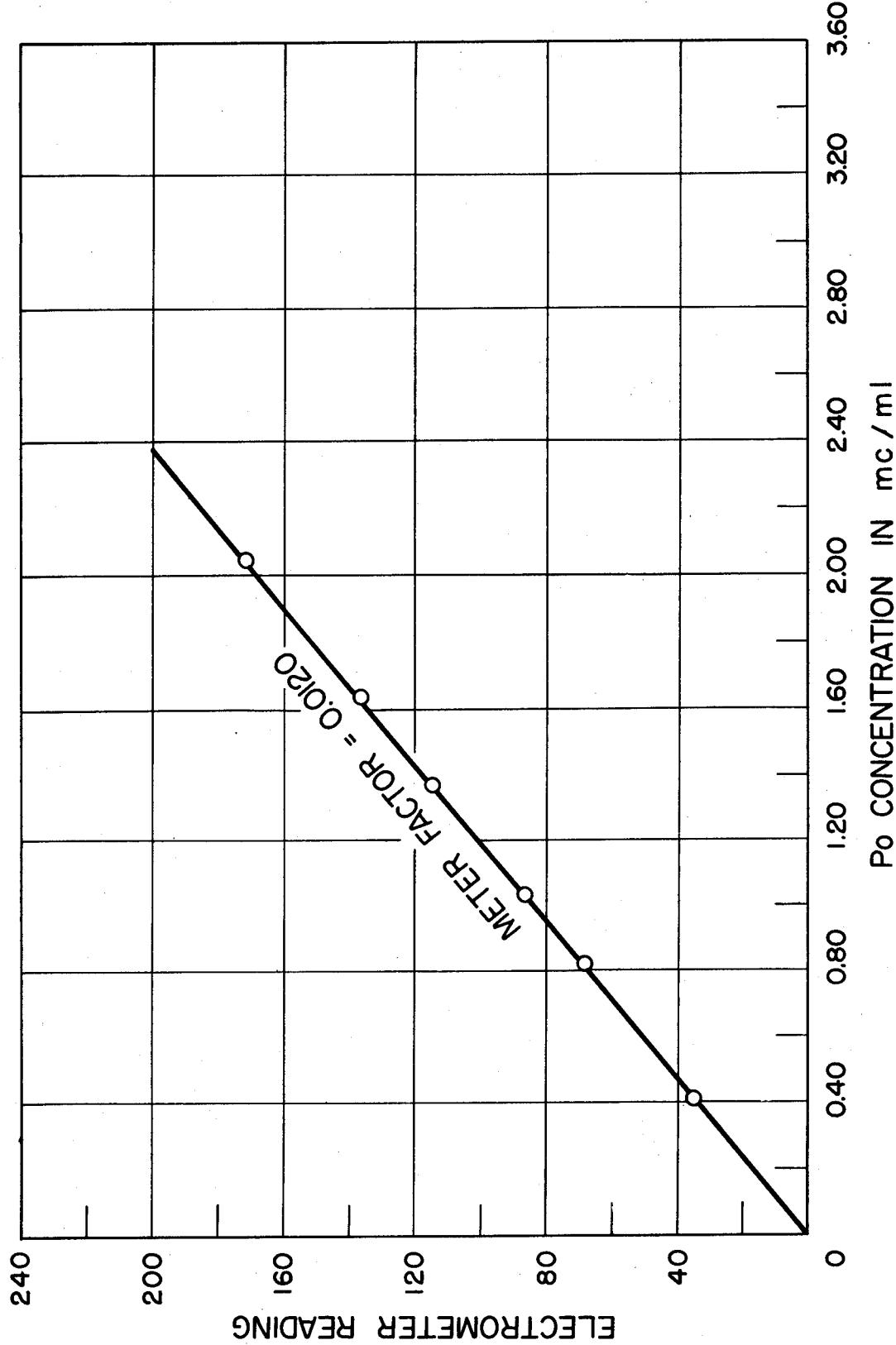


Fig. 5. Range 7 calibration curve.

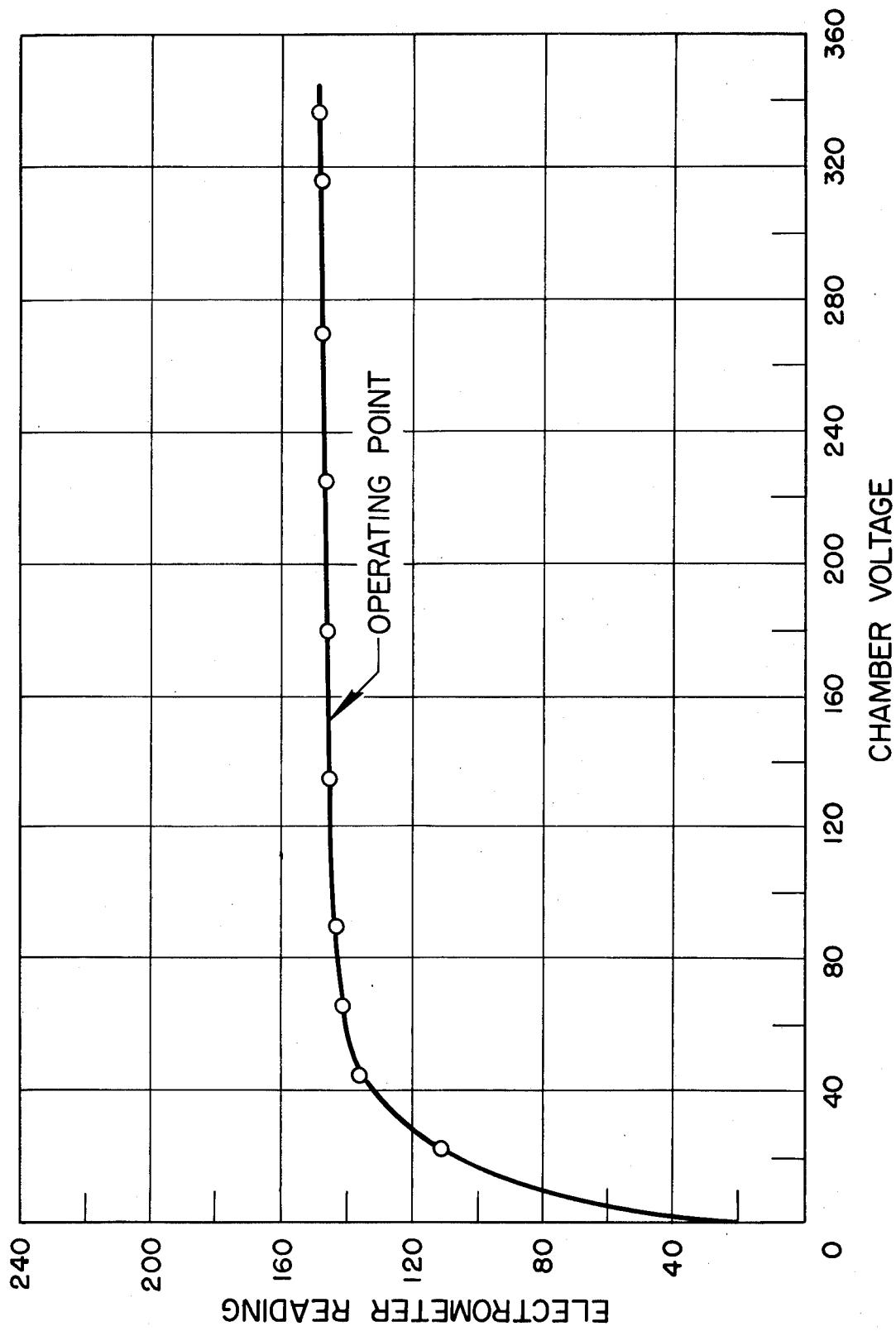


Fig. 6. Voltage plateau.

particles, the probe "sees" only those alphas which originate at a distance less than 0.003 cm from the probe window. Therefore, it is necessary to stir the solution to insure that the layer of solution immediately adjacent to the mica window is representative of the body of solution. Stirring is continued until the electrometer gives a constant maximum reading. With efficient stirring, this equilibrium is reached in about 1/2 min.

The polonium concentration is computed by multiplying the meter reading by the meter factor corresponding to the final range setting.

One problem in the operational use of the probe derives from contamination of the mica window. When the probe is to be used in a solution of low activity immediately after use in a fairly concentrated polonium solution, it is necessary to replace the liquid film on the mica window with an inert film. Simple washing with distilled water strongly reduces the activity but is not sufficient. Several decontamination methods were tried, including the formation of a silicone film on the mica, the use of wetting agents, and leaching of the probe with strong alkali and strong acid solutions. The most satisfactory procedure was a series of leachings in 6 N HCl interspersed with distilled water rinsings. When this technique was used, activity on the mica window was reduced from 1.0 mc/ml to less than 0.01 mc/ml in 10 min. No decontamination is required during electroplating operations in which the polonium concentration varies slowly and the bath is continuously stirred.

##### 5. Possible Errors in Solution Analyses

Inasmuch as the range of an alpha particle varies inversely with the stopping power of the medium, variations in the stopping power of the solutions will cause changes in the response of the electrometer and probe. To determine the magnitude of errors which might be expected from such variations, calculations were made of the range of polonium alphas in various solutions relative to the polonium alpha range in water. These data are presented in Fig. 7. Usually such errors will not be large, e.g., a change in nitric acid concentration from 2.0 N to 1.0 N will cause a change in response of less than 2.5 per cent. However, for very large changes in solution composition, rather large errors are possible. This possibility must be borne in mind if the probe is to be used to assay solutions of high acidity or high salt concentration. For the special case in which the concentration of the activity varies but the solution composition remains fairly constant, the probe will still prove accurate and useful. The response of the electrometer will be linear and directly proportional to the activity present so that only change in the meter factor is involved.

Another possible source of error is the presence of insoluble impurities. Since the probe is sensitive primarily to polonium atoms in solution, any phenomenon which tends to

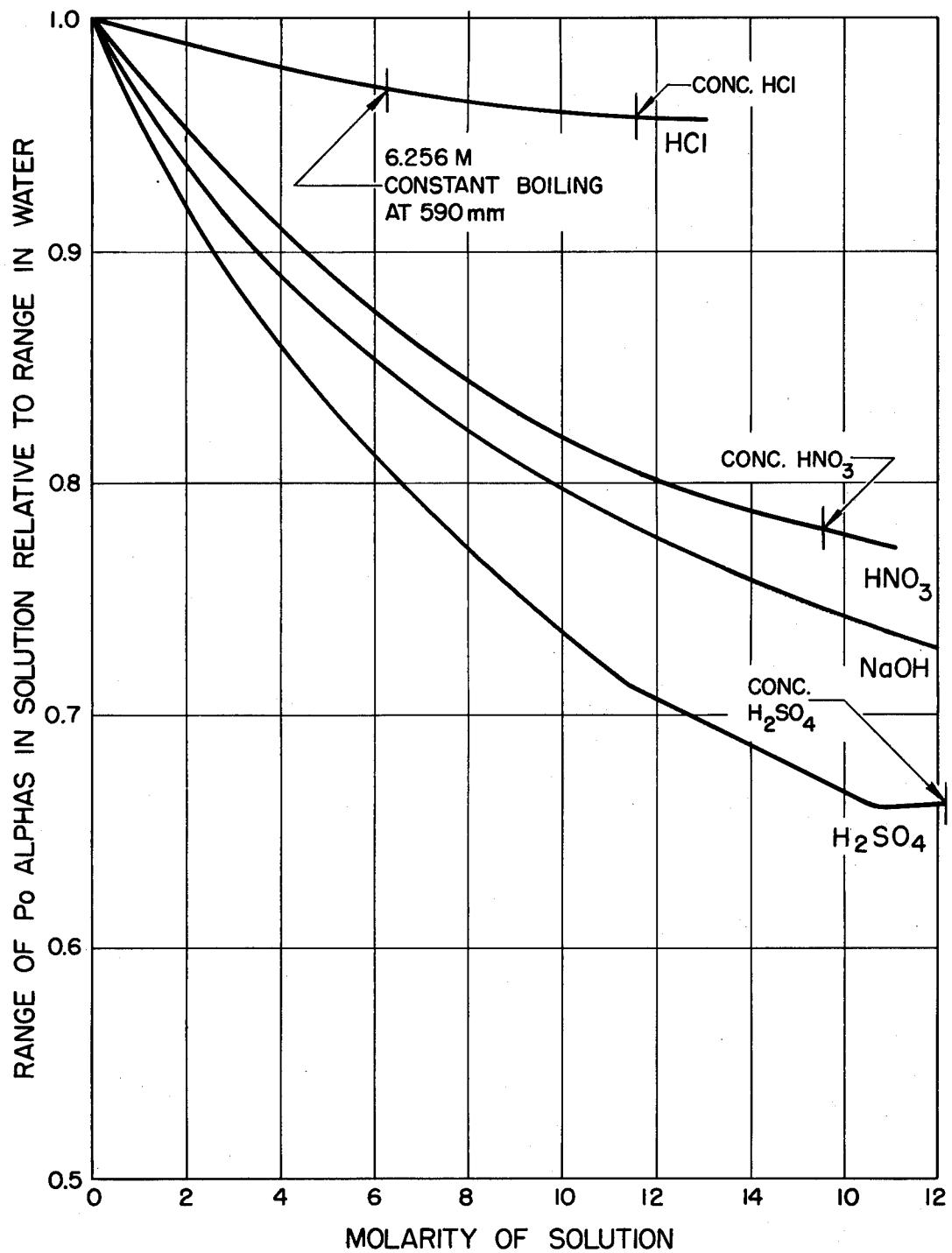


Fig. 7. Range of polonium alphas in solution.

remove activity from solution will result in low readings. This includes such effects as adsorption on the walls of the container and on insoluble impurities, formation of fine precipitates, and the formation of radiocolloids.

Minor variations in the temperature of the solution will be unimportant. The decrease in density due to an increase in temperature is compensated by an increase in the range of the alphas, and the amplifier response remains essentially constant.

#### 6. Conclusion

The construction and operational use of an immersion, probe-type alpha detector designed to permit direct determinations of the concentration of polonium in solution have been described.

Using polonium solutions of known concentrations, range factors for the various ranges of the electrometer amplifier were determined, as well as the response of the amplifier versus polonium concentrations over a complete range. Results indicate that the probe and electrometer have a linear response and will give reproducible values which are in error by only  $\pm 5$  per cent.

The outstanding features of the probe-type ionization chamber described are the simple construction, the ease of operation, and the fact that it gives an immediate and continuous indication of the levels of alpha activity in solution. It is, therefore, an effective instrument for the analysis of polonium in solution.

It is believed that the instrument described in this paper will prove to be very useful in a variety of problems involving solutions containing alpha emitters. If the probe construction were modified (perhaps a glass probe to which a mica window is attached by the use of powdered glass flux<sup>3</sup>) the instrument could be adapted for use with organic solutions.

#### 7. References

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2. F. M. Glass, "A Simple Low Drift Electrometer," Nucleonics 10, 36 (1952).
3. J. S. Donal, "Sealing Mica to Glass," Rev. Sci. Instr. 13, 266 (1942).